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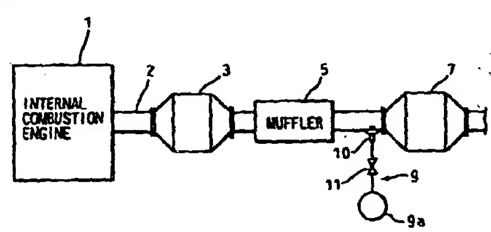
(54) A method for purifying combustion exhaust gas

(57) The method for purifying combustion exhaust gas according to the present invention utilizes a NH_3 decomposing catalyst. The NH_3 decomposing catalyst in the present invention is capable of converting substantially all of the NH_3 in the combustion exhaust gas to N_2 when the air-fuel ratio of the exhaust gas is lean and the temperature of the catalyst is within a predetermined optimum temperature range. Further, when the exhaust gas contains NO_x in addition to NH_3, the NH_3 decomposing catalyst is capable of reducing the NO_x in the optimum temperature range even though the air-fuel ratio of the exhaust gas is lean. In the present invention, the conditions of the exhaust gas containing NO_x are adjusted before it is fed to the NH_3 decomposing catalyst in such a manner that the temperature of the exhaust gas is within the optimum temperature range and the air-fuel ratio of the exhaust gas is lean. Further, NH_3 is added to the exhaust gas before it is fed to the NH_3 decomposing catalyst. Therefore, a lean air-fuel ratio exhaust gas, at a temperature within the optimum temperature range, which contains both the NO_x and NH_3 is fed to the NH_3 decomposing catalyst, and the NO_x, as well as the NH_3, in the exhaust gas is completely resolved by the NH_3 decomposing catalyst.

Fig. 1



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However, one problem arises when the process in the '917 publication is used for resolving HC, CO and NO_x components. The three-way reducing and oxidizing catalyst, though it has a high ability for reducing NO_x components in a reducing atmosphere, also converts a portion of NO_x components in the exhaust gas to NH_3 component (ammonia) in a reducing atmosphere. In the process in the '917 publication, since the rich air-fuel ratio exhaust gas is fed to the three-way reducing and oxidizing catalyst, the exhaust gas flows out from the three-way reducing and oxidizing catalyst contains a small amount of NH_3. This NH_3 in the exhaust gas is oxidized and again produces NO_x when the exhaust gas is fed to the oxidizing catalyst in an oxidizing atmosphere. Therefore, when the process in the '917 publication is used, it is difficult to resolve NO_x components in the exhaust gas completely, since NO_x components are produced by the oxidizing catalyst.

SUMMARY OF THE INVENTION

In view of the problems in the related art, the object of the present invention is to provide a process and a device for purifying a combustion exhaust gas which is capable of resolving substantially all of the HC, CO, and NO_x components as well as a NH_3 component in the exhaust gas while preventing NO_x from being produced by the oxidation of NH_3 contained in the exhaust gas.

The above object is achieved by a process for resolving an NH_3 component in a combustion exhaust gas by contacting a combustion exhaust gas containing an NH_3 component in an oxidizing atmosphere at a temperature within a predetermined range to an NH_3 decomposing catalyst which resolves the NH_3 component in an exhaust gas in an oxidizing atmosphere when the temperature of the catalyst is in the predetermined temperature range, converts the NH_3 component in the exhaust gas in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is high r than the predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than the predetermined temperature range.

The NH_3 decomposing catalyst is capable of resolving an NH_3 component in the exhaust gas in an oxidizing atmosphere when the temperature of the catalyst is within the predetermined temperature range. Therefore, by feeding the exhaust gas containing a NH_3 component to the NH_3 decomposing catalyst in an oxidizing atmosphere and at a temperature within the predetermined range, substantially all of the NH_3 component in the exhaust gas is resolved by the NH_3 decomposing catalyst without forming NO_x components.

According to another aspect of the present invention, there is provided a process for resolving pollutants in the exhaust gas of an internal combustion engine comprising, contacting an exhaust gas of an internal combustion engine in a reducing atmosphere with an NH_3 synthesizing catalyst which converts NO_x components in the exhaust gas, in a reducing atmosphere, to an NH_3 component, adjusting the conditions of the exhaust gas after it contacts the NH_3 synthesizing catalyst so that the exhaust gas is in an oxidising atmosphere and within a predetermined temperature range, and contacting the exhaust gas, after its atmosphere and temperature are adjusted, with an NH_3 decomposing catalyst which resolves an NH_3 component in the exhaust gas in an oxidizing atmosphere when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than the predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than the predetermined temperature range.

In this aspect of the invention, the exhaust gas from the internal combustion engine contacts the NH_3 synthesizing catalyst in an reducing atmosphere. Therefore, the NO_x component in the exhaust gas is converted to the NH_3 component. Further, after it contacts the NH_3 synthesizing catalyst, the exhaust gas contacts the NH_3 decomposing catalyst in an oxidizing atmosphere and at the temperature within the predetermined temperature range. Thus, substantially all of the NH_3 component produced by the NH_3 synthesizing catalyst is resolved by the NH_3 decomposing catalyst, and the exhaust gas flowing out from the NH_3 decomposing catalyst is completely free from the NO_x and NH_3 components.

Further, the NH_3 synthesizing catalyst in the present invention is capable of resolving most of pollutants in the exhaust gas when the exhaust gas is in a reducing atmosphere (i.e., when the oxygen concentration in the exhaust gas is low). For example, the pollutants such as HC, CO and NO in the exhaust gas are resolved by the NH_3 synthesizing catalyst by the following reactions when the exhaust gas is in a reducing atmosphere.

$$(CH_2)_m + (3m/2)O_2 \rightarrow mCO_2 + mH_2O \quad (1)$$

$$CO + (1/2)O_2 \rightarrow CO_2 \quad (2)$$

$$NO + CO \rightarrow (1/2)N_2 + CO_2 \quad (3)$$

and the NH_3 component in the exhaust gas is effectively resolved by the NH_3 decomposing catalyst even if the temperature of the exhaust gas at the outlet of the engine changes due to a change in the operating condition of the engine.

According to another aspect of the present invention, there is provided a process for resolving NO_x components in a combustion exhaust gas using an NH_3 decomposing catalyst which resolves NO_x component in the exhaust gas in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is within a predetermined temperature range, converts the NH_3 component in the exhaust gas in an oxidizing atmosphere when the temperature of the catalyst is higher than the predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than the predetermined temperature range comprising, supplying NH_3 to an exhaust gas from an internal combustion engine, and contacting the exhaust gas to the NH_3 decomposing catalyst in an oxidizing atmosphere and at the temperature within the predetermined temperature range.

In this aspect of the invention, the NH_3 decomposing catalyst resolves the NO_x components by reacting a NH_3 component with NO_x components in the exhaust gas in an oxidizing atmosphere when the temperature is within a predetermined temperature range. Since NH_3 is added to the exhaust gas before the exhaust gas is fed to the NH_3 decomposing catalyst in this aspect of the invention, the exhaust gas flowing into the NH_3 decomposing catalyst in oxidizing atmosphere and at the temperature within the predetermined temperature range contains NO_x and NH_3, the NO_x components in the exhaust gas react with the NH_3 component on the NH_3 decomposing catalyst and are resolved.

According to another aspect of the present invention, there is provided a device for resolving NO_x components in the exhaust gas of an internal combustion engine operated at an air-fuel ratio higher than the stoichiometric air-fuel ratio comprising an NH_3 decomposing catalyst disposed on an exhaust gas passage of an internal combustion engine, wher in the NH_3 decomposing catalyst resolves NO_x components in the exhaust gas flowing into the catalyst in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than the predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than the predetermined temperature range, an NH_3 supply means for supplying NH_3 to the exhaust gas flowing into the NH_3 decomposing catalyst, and temperature maintaining means for maintaining the t mperature of the exhaust gas flowing into the NH_3 decomposing catalyst within the predetermined temperature range regardless of change in the temperature of the exhaust gas discharged from the internal combustion engine.

In this aspect of the invention, the internal combustion engine is operated at a lean air-fuel ratio and the exhaust gas from the engine is in an oxidizing atmosphere. NH_3 is added to this exhaust gas before the exhaust gas flows into the NH_3 decomposing catalyst. Further, the temperature of the exhaust gas is maintained within the predetermined range. Therefore, the exhaust gas containing NO_x components and a NH_3 component is fed to the NH_3 decomposing catalyst in an oxidizing atmosphere and at the temperature within the predetermined temperature range, and the NO_x components in the exhaust gas reacts the NH_3 component at the NH_3 decomposing catalyst and resolved.

According to another aspect of the present invention, there is provided a device for resolving NO_x components in an exhaust gas of an internal combustion engine operated at an air-fuel ratio higher than the stoichiometric air-fuel ratio comprising a plurality of NH_3 decomposing catalysts disposed on the exhaust gas passage in series arrangement wherein each of the NH_3 decomposing catalysts resolves NO_x components in the exhaust gas flowing into the catalyst in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than the predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than the predetermined temperature range, an NH_3 supply means for supplying NH_3 selectively to the exhaust gas flowing into the respective NH_3 decomposing catalysts, temperature detecting means for detecting the temperature of the respective NH_3 decomposing catalysts, and selecting means for controlling the NH_3 supply means in such a manner that the NH_3 supply means supplies NH_3 to the exhaust gas flowing into the NH_3 decomposing catalyst(s) whose temperature is within the predetermined temperature range.

In this aspect of the invention, more than one NH_3 decomposing catalysts are disposed on the exhaust gas passage of the engine. Since the temperature of the exhaust gas becomes lower as the exhaust gas flows down the exhaust gas passage, the temperatures of the respective NH_3 decomposing catalysts vary in accordance with the location of the NH_3 decomposing catalysts. Therefore, even when the temperature of the exhaust gas changes due to change in the operating condition of the engine, some of the NH_3 decomposing catalysts always stay in the predetermined temperature range. A selecting means controls the NH_3 supply means so that NH_3 is supplied to the exhaust gas flowing into the NH_3 decomposing catalysts which has a temperature within the predetermined range. Therefore, exhaust gas in an oxidizing atm sphere containing NH_3 and NO_x is fed to the NH_3 decomposing catalyst which has the temperature within the predetermined range. Consequently, the NO_x component in the exhaust gas reacts the NH_3 component at the NH_3 decomposing catalyst and r solved.

In this aspect of the invention, the NH_3 decomposing catalyst is also capable of absorbing the NO_x components in the exhaust gas flowing into the NH_3 decomposing catalyst when the engine operating air-fuel ratio is higher than the stoichiometric air-fuel ratio. Thus, the NO_x components are not discharged to atmosphere when the engine operating air-fuel ratio is higher than the stoichiometric air-fuel ratio. When the engine operating air-fuel ratio becomes lower than the stoichiometric air-fuel ratio, a NH_3 component is produced by the NH_3 synthesizing catalyst upstream of the NH_3 decomposing catalyst. However, this NH_3 component is oxidized at the NH_3 decomposing catalyst by reacting the NO_x components absorbed by the NH_3 decomposing catalyst. Therefore, both the NO_x and NH_3 components are not discharged to atmosphere when the engine operating air-fuel ratio becomes lower than the stoichiometric air-fuel ratio.

BRIEF DESCRIPTION OF THE DRAWINGS

The present invention will be better understood from the description, as set forth hereinafter with reference to the accompanying drawings in which:

Fig. 1 is a drawing schematically illustrating an embodiment of the present invention when applied to an automobile engine;
Fig. 2 is a drawing schematically illustrating another embodiment of the present invention;
Fig. 3 is a drawing schematically illustrating another embodiment of the present invention;
Fig. 4 is a drawing schematically illustrating another embodiment of the present invention;
Fig. 5 is a drawing schematically illustrating another embodiment of the present invention;
Fig. 6 is a drawing showing the change in the characteristics of an NH_3 decomposing catalyst in accordance with the change in the temperature; and
Fig. 7 is a drawing schematically illustrating another embodiment of the present invention.

DESCRIPTION OF THE PREFERRED EMBODIMENT

In the embodiments explained hereinafter, NH_3 decomposing catalysts are used for resolving NO_x and NH_3 from a combustion exhaust gas. Therefore, an NH_3 decomposing catalyst is explained before explaining the respective embodiments.

The NH_3 decomposing catalyst in the embodiments of the present invention uses, for example, a honeycomb type substrate made of cordierite, and an alumina layer which act as a carrier for the catalyst is coated on the cell surface of the honeycomb substrate. On this carrier, at least one substance selected from elements belong to the fourth period or the eighth group in the periodic table of elements, such as copper (Cu), chrome (Cr), vanadium (V), titanium (Ti), iron (Fe), nickel (Ni), cobalt (Co), platinum (Pc), palladium (Pd), rhodium (Rh) and iridium (Ir) are carried as a catalyst.

The NH_3 decomposing catalyst is capable of converting all the NH_3 component in the exhaust gas flowing into the NH_3 decomposing catalyst to the N_2 component provided that the exhaust gas is in an oxidizing atmosphere and the temperature of the catalyst is within a specific temperature range as determined by the substance being used as the catalyst. Therefore, when the exhaust gas is an oxidizing atmosphere containing a NH_3 component and flows through the NH_3 decomposing catalyst in this temperature range, the NH_3 component in the exhaust gas is almost completely resolved, and the exhaust gas flows out from the NH_3 decomposing catalyst contains no NH_3 component. In the explanation below, this temperature range in which the NH_3 decomposing catalyst can resolve all the NH_3 component in the exhaust gas is called an optimum temperature range.

When the temperature of the NH_3 decomposing catalyst is higher than the optimum temperature range, the NH_3 component in the exhaust gas flowing into the NH_3 decomposing catalyst is oxidized by the NH_3 decomposing catalyst and NO_x components are produced.

Namely, when the temperature of the NH_3 decomposing catalyst is higher than the optimum temperature range, the oxidizing reaction of the NH_3 component, i.e.,

$$4NH_3 + 7O_2 \rightarrow 4NO_2 + 6H_2O, \text{ and/or } 4NH_3 + 5O_2 \rightarrow 4NO + 6H_2O$$

become dominant on the NH_3 decomposing catalyst, and the amount of NO_x components (mainly NO and NO_2) in the exhaust gas flowing out from the NH_3 decomposing catalyst increases.

Further, when the temperature of the NH_3 decomposing catalyst is lower than the optimum temperature range, the oxidizing reaction of the NH_3 component becomes lower, and the amount of the NH_3 component in the exhaust gas flowing out from the NH_3 decomposing catalyst increases.

Fig. 6 schematically illustrates the change in the characteristics of the NH_3 decomposing catalyst in accordance with the change in the temperature. Fig. 6 shows the change in the concentration of the NH_3 and NO_x components in the exhaust gas flowing out from the NH_3 decomposing catalyst in accordance with the temperature of the NH_3 decomposing catalyst when the exhaust gas flowing into the NH_3 decomposing catalyst is in an oxidizing atmosphere and the

NO_x produced by the oxidizing reactions flows out from the NH_3 decomposing catalyst without being reduced by the denitrating reactions.

On the other hand, when the temperature of NH_3 decomposing catalyst is below the optimum temperature range, the oxidizing reactions hardly occur due to the low temperature. This causes the NH_3 in the exhaust gas passes through the NH_3 decomposing catalyst without being oxidized by the NO_x due to the shortage of the NO_x in the exhaust gas.

As explained above, the optimum temperature range of the NH_3 decomposing catalyst is a temperature range in which the oxidizing reactions of the NH_3 and the denitrating reactions of the NO_x balance each other in such a manner that the NO_x produced by the oxidation of the NH_3 immediately reacts with NH_3 in the exhaust gas without causing any surplus NO_x and NH_3. Consequently, the optimum temperature range of the NH_3 decomposing catalyst is determined by the oxidizing ability of the catalyst and its temperature dependency. Therefore, when the catalyst component having high oxidizing ability, such as platinum (Pt), is used, the optimum temperature range becomes lower than that when the catalyst component having relatively low oxidizing ability, such as chrome (Cr) is used.

As explained above, though the mechanism of the phenomenon is not completely clarified, the NH_3 decomposing catalyst actually converts all of the NH_3 in the exhaust gas in an oxidizing atmosphere when the temperature is within the optimum temperature range. Further, when the NH_3 decomposing catalyst is used in the optimum temperature range the following facts were found in connection with the above phenomenon:

(a) When the exhaust gas flowing into the NH_3 decomposing catalyst is in an oxidizing atmosphere, i.e., when the air-fuel ratio of the exhaust gas is lean compared to the stoichiometric air-fuel ratio, substantially all of the NH_3 in the exhaust gas is converted to N_2 without producing any NO_x. This occurs when the exhaust gas is in an oxidizing atmosphere (a lean air-fuel ratio), but regardless of the degree of leanness of air-fuel ratio of the exhaust gas. (In this specification, an air-fuel ratio of the exhaust gas at a certain point is defined by a ratio of the air and the fuel which are supplied to the combustion chambers or exhaust passages upstream of the point. Therefore, when no air or fuel is supplied in the exhaust passages upstream of the considered point, the air-fuel ratio of the exhaust gas at the point becomes the same as the air-fuel ratio of the air-fuel mixture supplied to the combustion chambers).

(b) When the exhaust gas flowing into the NH_3 decomposing catalyst contains NO_x in addition to NH_3, all of the NO_x in the exhaust gas as well as the NH_3 is converted to N_2, and the concentration of the NO_x components in the exhaust gas becomes zero. In this case, the ratio of the concentrations of the NO_x components and the NH_3 component is not necessarily stoichiometrical for the denitrating reactions (i.e., 4:3, or 1:1). It is only required that the exhaust gas contains an amount of NH_3 more than the amount required for reducing the NO_x (NO_2 and NO) in the exhaust gas. As explained above, since the surplus NH_3 in the exhaust gas is all converted to N_2 when the exhaust gas is in an oxidizing atmosphere, no surplus NH_3 is contained in the exhaust gas flowing out from the NH_3 decomposing catalyst even in this case.

(c) When the exhaust gas flowing into the NH_3 decomposing catalyst contains HC and CO components, all of the HC and CO components are oxidized by the NH_3 decomposing catalyst, provided that the air-fuel ratio of the exhaust gas is lean compared to the stoichiometric air-fuel ratio, and no HC and CO components are contained in the exhaust gas flowing out from the NH_3 decomposing catalyst.

However, when the exhaust gas flowing into the NH_3 decomposing catalyst contains both the NH_3 and NO_x, it was found that the temperature region IV in Fig. 6, i.e., the temperature region in which the concentration of NO_x components in the outflow exhaust gas increases as the temperature of the catalyst increases, moves to the lower temperature side compared to that when the exhaust gas flowing into the NH_3 decomposing catalyst contains only the NH_3 components. This is because, when the exhaust gas contains NO_x in addition to NH_3, the NO_x in the inflow exhaust gas in addition to the NO_x produced by the oxidizing reaction of NH_3 must be reduced by the NH_3 in the exhaust gas. Consequently, the shortage of the NH_3 is apt to occur in the relatively low temperature region. Therefore, when the exhaust gas contains both the NH_3 and the NO_x, the optimum temperature range of the NH_3 decomposing catalyst becomes narrower.

In relation to above (b), a conventional denitrating catalyst, such as a vanadia-titania (V_2O_5-TiO_2) type catalyst also has a capability for resolving NH_3 and NO_x in the exhaust gas under a certain conditions. However, in case of the conventional denitrating catalyst, the amounts of NH_3 and NO_x components must be strictly stoichiometrical in order to react NH_3 with NO_x without causing any surplus NH_3 and NO_x. Namely, when both the NO_2 and NO are contained in the exhaust gas, the amount (moles) of the NH_3 in the exhaust gas must be strictly equal to the total of the moles of NO_2 in the exhaust gas multiplied by 3/4 and the moles of NO in the exhaust gas in order to react NH_3 and NO_x without causing any surplus NH_3 and NO_x. However, in case of the NH_3 decomposing catalyst in the embodiments of the present invention, if the amount of the NH_3 is more than the stoichiometrical compared to the amount of NO_x, and if the air-fuel ratio of the exhaust gas is lean, all of the NH_3 and NO_x are converted to N_2 without causing any surplus NH_3 and NO_x. This is an important difference between the NH_3 decomposing catalyst in the present invention and the conventional denitrating catalyst.

In this embodiment, the NH_3 decomposing catalyst 7 is disposed on the exhaust gas passage downstream of the three-way reducing and oxidizing catalyst 3 in order to resolve the NH_3 formed at the three-way reducing and oxidizing catalyst 3. Namely, the exhaust gas flows out from the three-way reducing and oxidizing catalyst 3 flows down the exhaust gas passage 2 and passes through the muffler 5. Further, air is added to the exhaust gas before it flows into the NH_3 decomposing catalyst 7 by the secondary air supply unit 9.

Usually, the temperature of the exhaust gas at the outlet of the engine 1 is above the optimum temperature range of the NH_3 decomposing catalyst 7. However, the temperature of the exhaust gas becomes lower as the exhaust gas flows down the exhaust gas passage due to the heat dissipation through the wall of the exhaust gas passage 2. Further, when it passes through the muffler 5, the exhaust gas is cooled by the muffler 5. Therefore, the temperature of th exhaust gas when it reaches the NH_3 decomposing catalyst 7 is lower than the temperature at the outlet of the engin 1. In this embodiment, the length of the exhaust gas passage between the engine 1 and the NH_3 decomposing catalyst 7 as well as the capacity of the muffler 5 is determined in such a manner that the temperature of the exhaust gas at th inlet of the NH_3 decomposing catalyst 7 falls in the optimum temperature range even when the engine 1 is operated at the maximum exhaust temperature conditions. By this arrangement, the temperature of the exhaust gas flowing into the NH_3 decomposing catalyst 7, and the temperature of the NH_3 decomposing catalyst 7 accordingly, is always maintained within the optimum temperature range regardless of the change in the operating conditions of the engine 1.

Further, the air-fuel ratio of the exhaust gas flowing into the NH_3 decomposing catalyst 7 is adjusted by supplying air from the secondary air supply unit 9 so that the air-fuel ratio of the exhaust gas becomes lean compared to the stoichiometric air-fuel ratio. As explained before, when the exhaust gas flowing into the NH_3 decomposing catalyst 7 is in an oxidizing atmosphere (i.e., at a lean air-fuel ratio), the NH_3 decomposing catalyst 7 is capable of resolving substantially all of the NH_3 in the exhaust gas regardless of the degree of the leanness of the air-fuel ratio of the exhaust gas. Therefore, it is not necessary to control the amount of air supplied from the secondary air supply unit 9 in order to strictly control the air-fuel ratio of the exhaust gas flowing into the NH_3 decomposing catalyst 7 in this embodiment. The amount of the air supplied from the secondary air supply unit 9 is set at an amount which can keep the air-fuel ratio of the exhaust gas flowing into the NH_3 decomposing catalyst 7 on a lean air-fuel ratio side compared to the stoichiometric air-fuel ratio even when the operating air-fuel ratio of the engine 1 fluctuates to the rich air-fuel ratio side compared to the stoichiometric air-fuel ratio.

By the arrangement explained above, the exhaust gas supplied to the NH_3 decomposing catalyst 7 is always an oxidizing atmosphere and has a temperature within the optimum temperature range. Therefore, even if NH_3 is formed at the three-way reducing and oxidizing catalyst 3, NH_3 is completely resolved by the NH_3 decomposing catalyst 7. Further, when the air-fuel ratio of the exhaust gas becomes rich, the capability of the three-way reducing and oxidizing catalyst 3 for resolving HC and CO component in the exhaust gas decreases. However, according to the present embodiment, HC and CO components passing through the three-way reducing and oxidizing catalyst 3 are also resolved by the NH_3 decomposing catalyst 7. Therefore, according to the present embodiment, an increase in the emission of HC, CO and NH_3 components when the operating air-fuel ratio of the engine fluctuates to rich side can be prevented.

Though the temperature of the exhaust gas is adjusted by the heat dissipation from the wall of the exhaust gas passage 2 and the cooling by the muffler 5 in this embodiment, other means for adjusting the temperature of the exhaust gas may be used. For example, heat radiation fins may be disposed outer wall of the exhaust gas passage 2 instead of, or in addition to the muffler 5, or alternatively, the wall of the exhaust gas passage 2 may be water-cooled to increase the cooling capacity of the exhaust gas passage 2. Further, the amount of the air supplied from the secondary air supply unit 9 may be changed in accordance with the temperature of the exhaust gas to maintain the temperature of the exhaust gas flowing into the NH_3 decomposing catalyst 7 within the optimum temperature range by using a flow control valve instead of the shut off valve 11 in this embodiment. Further, the flow amount of the cooling water for cooling the exhaust gas passage wall, or the amount of the air supplied from the secondary air supply unit may be feedback controlled in accordance with an output signal of a temperature sensor detecting the catalyst bed of the NH_3 decomposing catalyst 7 in such a manner that the temperature of the NH_3 decomposing catalyst is maintained within the optimum temperature range.

In the above embodiment, if the operating air-fuel ratio of the engine 1 fluctuates to a lean air-fuel ratio side compared to the stoichiometric air-fuel ratio, NH_3 is not formed at the three-way reducing and oxidizing catalyst 3, and the capability of the three-way reducing and oxidizing catalyst for reducing NO_x also decreases. In this case, an exhaust gas which contains NO_x, but does not contain NH_3 flows into the NH_3 decomposing catalyst 7. This causes the NO_x in the exhaust gas to pass through the NH_3 decomposing catalyst 7 without being reduced. Therefore, to prevent the emission of the NO_x when the operating air-fuel ratio becomes lean, a NO_x absorbent which is capable of absorbing NO_x in the exhaust gas of a lean air-fuel ratio, or a NO_x reducing catalyst which has a capability for selectively reducing the NO_x in the exhaust gas even in an oxidizing atmosphere may be disposed on the exhaust gas passage 2 between the three-way reducing and oxidizing catalyst 3 and the NH_3 decomposing catalyst 7. Fig. 7 shows an embodiment of the present invention in which a NO_x absorbent or a NO_x reducing catalyst is disposed on the exhaust gas passage 2 between the three-way reducing and oxidizing catalyst 3 and the NH_3 decomposing catalyst 7. In Fig. 7, same reference

the form of nitric acid ions NO_3^-. Thus, NO_x in the exhaust gas is absorbed by the NO_x absorbent 8 when the air-fuel ratio of the exhaust gas is lean.

On the other hand, when the oxygen concentration in the exhaust gas becomes low, i.e., when the air-fuel ratio of the exhaust gas becomes rich, the production of NO_2 on the surface of the platinum (Pt) is lowered and the reaction proceeds in an inverse direction

$$(NO_3^- \rightarrow NO_2),$$

and thus nitric acid ions NO_3^- in the absorbent are released in the form of NO_2 from the NO_x absorbent 8.

In this case, if the reducing substance such as NH_3 and CO, or the substance such as HC and CO_2 exist in the exhaust gas, the released NO_x is reduced on the platinum Pt by these components. Namely, the NO_x absorbent 8 performs the absorbing and releasing operation of the NO_x in the exhaust gas in which the NO_x in the exhaust gas is absorbed by the NO_x absorbent when the air-fuel ratio of the exhaust gas is lean and released from the NO_x absorbent when the air-fuel ratio of the exhaust gas becomes rich.

As explained in Fig. 6, the NH_3 decomposing catalyst 7 converts the NH_3 in the exhaust gas to NO_x when the temperature becomes higher than the optimum temperature range. Therefore, if the exhaust gas temperature at the outlet of the engine 1 becomes very high in an extreme operating condition of the engine 1, the temperature maintaining means such as the muffler 5 might be insufficient to lower the exhaust gas temperature to the optimum temperature range of the NH_3 decomposing catalyst 7. In such a case, since the temperature of the exhaust gas flowing into the NH_3 decomposing catalyst 7 exceeds the optimum temperature range, NO_x is formed at the NH_3 decomposing catalyst 7 and is discharged to atmosphere.

In this embodiment, the NO_x absorbent 8 is disposed on the exhaust gas passage 2 downstream of the NH_3 decomposing catalyst 7 to prevent the emission of the NO_x in the extremely high exhaust gas temperature conditions. Since the NO_x absorbent 8 absorbs the NO_x in the exhaust gas in an oxidizing atmosphere, the NO_x formed at the NH_3 decomposing catalyst 7 in the extremely high exhaust gas temperature condition is absorbed by the NO_x absorbent 8 and the emission of the NO_x to the atmosphere does not occur.

As explained above, according to the present embodiment, the exhaust emission can be always kept low even when the exhaust gas temperature becomes extremely high.

Though the NO_x absorbent 8 in the above embodiment is disposed on the exhaust gas passage 2 separately, it is possible to give the absorbing and releasing capability of NO_x to the substrate of the NH_3 decomposing catalyst. This is accomplished by attaching the NO_x absorbing substances such as alkali metals and alkali-earth metals to the substrate of the NH_3 decomposing catalyst 7 in addition to the catalytic components.

Next, another embodiment of the present invention is explained. In this embodiment, an NO_x reducing catalyst is disposed on the exhaust gas passage downstream of the NH_3 decomposing catalyst 7 instead of the NO_x absorbent 8 in Fig. 2. Other constructions of this embodiment are the same as those in Fig. 2. Therefore, illustration of this embodiment by drawings is omitted.

The NO_x reducing catalyst in this embodiment has a substrate made of, for example, zeolite ZSM-5, and metals such as copper (Cu) and iron (Fe) are attached to the substrate by an ion exchange method. Alternatively, a substrate made of zeolite such as mordenite and precious metal such as platinum (Pt) attached thereon can also be used as the NO_x reducing catalyst. The NO_x reducing catalyst traps NH_3, HC and CO components in the exhaust gas in the pores of the porous zeolite, and selectively reduces the NO_x in the exhaust gas using these trapped components even in an oxidizing atmosphere.

In this embodiment, the NO_x reducing catalyst disposed at downstream of the NH_3 decomposing catalyst 7 in Fig. 2 traps the NH_3 component in the exhaust gas which passes through the NH_3 decomposing catalyst 7 when the exhaust gas temperature is below the optimum temperature range as well as the HC and CO components in the exhaust gas. Further, when the exhaust gas temperature is above the optimum temperature range, the NO_x reducing catalyst selectively reduces the NO_x formed at the NH_3 decomposing catalyst 7 using the trapped NH_3, HC and CO comp nents. Therefore, according to the present embodiment, the NO_x formed at the NH_3 decomposing catalyst in the high exhaust gas temperature conditions is not emitted to atmosphere and the exhaust emissions are always maintained at low level regardless of the change in the operating conditions of the engine.

Next another embodiment of the present invention is explained with reference to Fig. 3. In Fig. 3, the same referenc numerals as those in Fig. 1 designate the same elements.

In this embodiment, means for cooling the exhaust gas such as the muffler 5 in Fig. 1 is not used. Further, a plurality of the NH_3 decomposing catalyst are disposed on the exhaust gas passage 2 downstream of the three-way reducing and oxidizing catalyst 3 (Fig. 3 shows the case in which three NH_3 decomposing catalyst 7a to 7c are used). Also, the secondary air supply unit 9 in this embodiment has a plurality of the nozzles 10a to 10c which are each disposed, in the exhaust gas passage 2, at the inlet of the respective NH_3 decomposing catalysts 7a to 7c. Reference numeral 30 in Fig. 3 designates a control circuit of the engine 1. The control circuit 30 may be comprises, for example, a micro-computer of known type and performs basic controls of the engine 1 such as a fuel injection control and an ignition timing control.

posed on the exhaust gas passage 42. However, a three-way reducing and oxidizing catalyst 3 as shown in Fig. 5 is not provided in this embodiment. Further, instead of the secondary air supply unit 9 in Fig. 1, an NH_3 supply unit 49 which comprises a nozzle 50 and a shut off valve 51 which are similar to those in Fig. 1 and a NH_3 supply source 49a such as a bottle containing gaseous or liquid NH_3 is provided in this embodiment.

In this embodiment, the exhaust gas in an oxidizing atmosphere from the lean burn engine 41 is cooled by the muffler 45 and flows into the NH_3 decomposing catalyst 47 at the temperature within the optimum temperature range. Further, NH_3 is added to the exhaust gas at the portion upstream of the NH_3 decomposing catalyst 47. The exhaust gas from the lean burn engine 41 contains a relatively large amount of NO_x. Thus, the exhaust gas flowing into the NH_3 decomposing catalyst is adjusted so that it becomes an oxidizing atmosphere and a temperature within the optimum temperature range. Further, the exhaust gas contains NO_x and NH_3. Therefore, both the NO_x and the NH_3 in the exhaust gas are completely resolved by the NH_3 decomposing catalyst 47.

As explained before, the capability of the three-way reducing and oxidizing catalyst for reducing the NO_x becomes very small when the exhaust gas is in an oxidizing atmosphere. Therefore, it is difficult to resolve the NO_x in the exhaust gas of the lean burn engine using the three-way reducing and oxidizing catalyst. However, according to the present invention, the NO_x in the exhaust gas from the lean burn engine can be completely resolved by the NH_3 decomposing catalyst 47.

Further, the method for resolving NO_x in the exhaust gas of in an oxidizing atmosphere by adding an NH_3 component to the exhaust gas and using a conventional vanadia-titania (V_2O_5-TiO_2) type denitrating catalyst is known in the art. In the above conventional method, NO_x and NH_3 are converted to N_2 and H_2O by the denitrating reactions ($8NH_3 + 6NO_2 \rightarrow 12H_2O + 7N_2$ and/or $4NH_3 + 4NO_2 + O_2 \rightarrow 6H_2O + 4N_2$). However, in the conventional method, the amount of NO_x components and the NH_3 component must be adjusted so that the ratio of the moles of NO_x and NH_3 are strictly stoichiometrical (i.e., 4:3 or 1:1) in order to react NO_x with NH_3 without causing any surplus NO_x and NH_3, as explained before.

However, in the actual operation of the engine, the concentration of the NO_x components in the exhaust gas varies widely in accordance with the operating condition of the engine, and it is difficult to control the amount of the NH_3 added to the exhaust gas in accordance with the concentration of the NO_x in the exhaust gas. Therefore, if the conventional method is used for an actual engine, NO_x or NH_3 is emitted to the atmosphere in some cases.

However, in case of the NH_3 decomposing catalyst 47 in this embodiment, it is not required to strictly control the amount of the NH_3 added to the exhaust gas since the NH_3 decomposing catalyst can convert all of the NH_3 in the exhaust gas as long as the exhaust gas is in an oxidizing atmosphere. In this embodiment, it is only required that the amount of the NH_3 added to the exhaust gas is sufficiently large to reduce all of the NO_x in the exhaust gas while maintaining the exhaust gas in an oxidizing atmosphere. Therefore, according to the present embodiment, the NO_x in the exhaust gas of the lean burn engine can be completely resolved by a simple control of the NH_3 supply unit 49 while preventing the emission of NH_3.

Fig. 5 shows another embodiment of the present invention which is similar to the embodiment in Fig. 3. In this embodiment, a plurality of the NH_3 decomposing catalysts (47a to 47c in Fig. 5) and NH_3 supply nozzles (50a to 50c in Fig. 5) are disposed on the exhaust gas passage 42. Similarly to the embodiment in Fig. 3, the control circuit 30 selects the NH_3 decomposing catalyst which is in the optimum temperature range according to the operating conditions of the engine 41, and supplies NH_3, to those NH_3 decomposing catalysts through the NH_3 nozzle located at the inlet of those NH_3 decomposing catalysts. Since the operation of the present embodiment is sub stantially the same as the operation of the embodiment in Fig. 3, a detailed explanation is not given here.

From the description set forth above, it will be understood that, according to the present invention, the NH_3 and NO_x components contained in the combustion exhaust gas can be resolved effectively by the NH_3 decomposing catalyst. Though the present invention is explained using the embodiments in which the present invention is applied to the internal combustion engine, the application of the present invention is not limited to the internal combustion engine. The present invention can be also applied to resolve the NH_3 and NO_x components in the exhaust gas discharged from the combustion devices other than the internal combustion engine. Namely, the present invention can be also applied to, for example, boilers, furnaces etc., which emit combustion exhaust gases.

The method for purifying combustion exhaust gas according to the present invention utilizes a NH_3 decomposing catalyst. The NH_3 decomposing catalyst in the present invention is capable of converting substantially all of the NH_3 in the combustion exhaust gas to N_2 when the air-fuel ratio of the exhaust gas is lean and the temperature of the catalyst is within a predetermined optimum temperature range. Further, when the exhaust gas contains NO_x in addition to NH_3, the NH_3 decomposing catalyst is capable of reducing the NO_x in the optimum temperature range even though the air-fuel ratio of the exhaust gas is lean. In the present invention, the conditions of the exhaust gas containing NO_x are adjusted before it is fed to the NH_3 decomposing catalyst in such a manner that the temperature of the exhaust gas is within the optimum temperature range and the air-fuel ratio of the exhaust gas is lean. Further, NH_3 is added to the exhaust gas before it is fed to the NH_3 decomposing catalyst. Therefore, a lean air-fuel ratio exhaust gas, at a temperature within the optimum temperature range, which contains both the NO_x and NH_3 is fed to the NH_3 decomposing catalyst, and the NO_x, as well as the NH_3, in the exhaust gas is completely resolved by the NH_3 decomposing catalyst.

9. A device according to claim 7, further comprising an NO_x reducing catalyst which selectively reduces NO_x components in the exhaust gas in an oxidizing atmosphere and is disposed on the exhaust gas passage downstream of said NH_3 decomposing catalyst.

10. A device for resolving an NH_3 component contained in the exhaust gas of an internal combustion engine comprising:

a plurality of NH_3 decomposing catalysts disposed in an exhaust gas passage of an internal combustion engine in series arrangement, each of said NH_3 decomposing catalysts resolving an NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than said predetermined temperature range, and allowing the NH_3 component in an exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than said predetermined temperature range;

oxygen supply means for supplying oxygen selectively to the exhaust gas flowing into the respective NH_3 decomposing catalysts;

temperature detecting means for detecting the temperature of the respective NH_3 decomposing catalysts: and

selecting means for controlling said oxygen supply means in such a manner that the oxygen supply means supplies oxygen to the exhaust gas flowing into the NH_3 decomposing catalyst(s) whose temperature is within said predetermined temperature range.

11. A process for resolving NO_x components from a combustion exhaust gas using an NH_3 decomposing catalyst which resolves NO_x component in the exhaust gas in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is within a predetermined temperature range, converts the NH_3 component in the exhaust gas in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than said predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than said predetermined temperature range comprising:

supplying NH_3 to an combustion exhaust gas; and

contacting the exhaust gas with the NH_3 decomposing catalyst in an oxidizing atmosphere and at the temperature within said predetermined temperature range.

12. A device for resolving NO_x components in the exhaust gas of an internal combustion engine operated at an air-fuel ratio higher than the stoichiometric air-fuel ratio comprising:

an NH_3 decomposing catalyst disposed on an exhaust gas passage of an internal combustion engine, said NH_3 decomposing catalyst resolves NO_x components in the exhaust gas flowing into the catalyst in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than said predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than said predetermined temperature range;

NH_3 supply means for supplying NH_3 to the exhaust gas flowing into said NH_3 decomposing catalyst; and

temperature maintaining means for maintaining the temperature of the exhaust gas flowing into said NH_3 decomposing catalyst within said predetermined temperature range regardless of a change in the temperature of th exhaust gas discharged from the internal combustion engine.

13. A device for resolving NO_x components in an exhaust gas of an internal combustion engine operated at an air-fuel ratio higher than the stoichiometric air-fuel ratio comprising:

a plurality of NH_3 decomposing catalysts disposed in the exhaust gas passage in series arrangement, each of said NH_3 decomposing catalysts resolving NO_x components in the exhaust gas flowing into the catalyst in an oxidizing atmosphere under the presence of an NH_3 component when the temperature of the catalyst is in a predetermined temperature range, converts the NH_3 component in the exhaust gas flowing into the catalyst in an oxidizing atmosphere to NO_x components when the temperature of the catalyst is higher than said predetermined temperature range, and allows the NH_3 component in the exhaust gas in an oxidizing atmosphere to pass through the catalyst when the temperature of the catalyst is lower than said predetermined temperature range;

NH_3 supply means for supplying NH_3 selectively to the exhaust gas flowing into the respective NH_3 decomposing catalysts;

temperature detecting means for detecting the temperature of the respective NH_3 decomposing catalysts; and

21. A device according to claim 10, wherein said NH_3 decomposing catalyst contains an NH_3 adsorbing component which adsorbs an NH_3 component in the exhaust gas.

22. A device according to claim 12, wherein said NH_3 decomposing catalyst contains an NH_3 adsorbing component which adsorbs an NH_3 component in the exhaust gas.

23. A device according to claim 13, wherein said NH_3 decomposing catalyst contains an NH_3 adsorbing component which adsorbs an NH_3 component in the exhaust gas.

24. A process according to claim 17, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

25. A process according to claim 18, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

26. A process according to claim 19, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

27. A device according to claim 20, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

28. A device according to claim 21, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

29. A device according to claim 22, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

30. A device according to claim 23, wherein said NH_3 adsorbing component adsorbs an NH_3 component in the exhaust gas when the temperature is lower than said predetermined temperature range.

31. A device according to claim 20, wherein said NH_3 adsorbing component comprises an acidic inorganic substance.

32. A device according to claim 21, wherein said NH_3 adsorbing component comprises an acidic inorganic substance.

33. A device according to claim 22, wherein said NH_3 adsorbing component comprises an acidic inorganic substance.

34. A device according to claim 23, wherein said NH_3 adsorbing component comprises an acidic inorganic substance.

35. A device according to claim 20, wherein said NH_3 adsorbing component comprises at least one substance selected from zeolite, silica (SiO_2), titania (TiO_2), silica-alumina (SiO_2-Al_2O_3) copper (Cu), cobalt (Co), nickel (Ni) and iron (Fe).

36. A device according to claim 21, wherein said NH_3 adsorbing component comprises at least one substance selected from zeolite, silica (SiO_2), titania (TiO_2), silica-alumina (SiO,-Al_2O_3) copper (Cu), cobalt (Co) , nickel (Ni) and iron (Fe).

37. A device according to claim 22, wherein said NH_3 adsorbing component comprises at least one substance selected from zeolite, silica (SiO_2), titania (TiO_2), silica-alumina (SiO_2-Al_2O_3) copper (Cu), cobalt (Co), nickel (Ni) and iron (Fe).

38. A device according to claim 23, wherein said NH_3 adsorbing component comprises at least one substance selected from zeolite, silica (SiO_2), titania (TiO_2), silica-alumina (SiO_2-Al_2O_3) copper (Cu), cobalt (Co), nickel (Ni) and iron (Fe).

Fig. 2

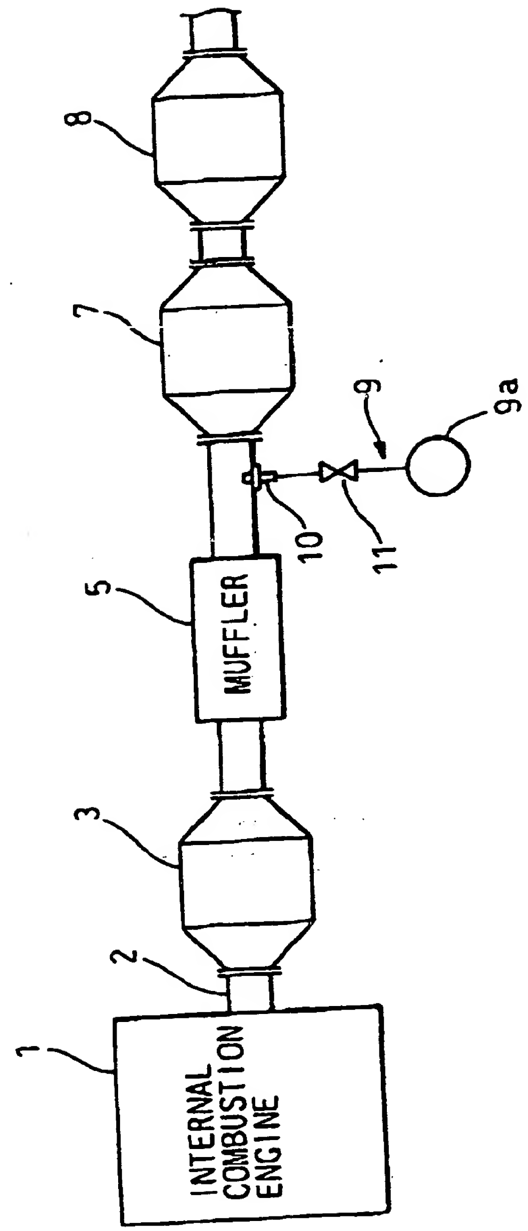


Fig.4

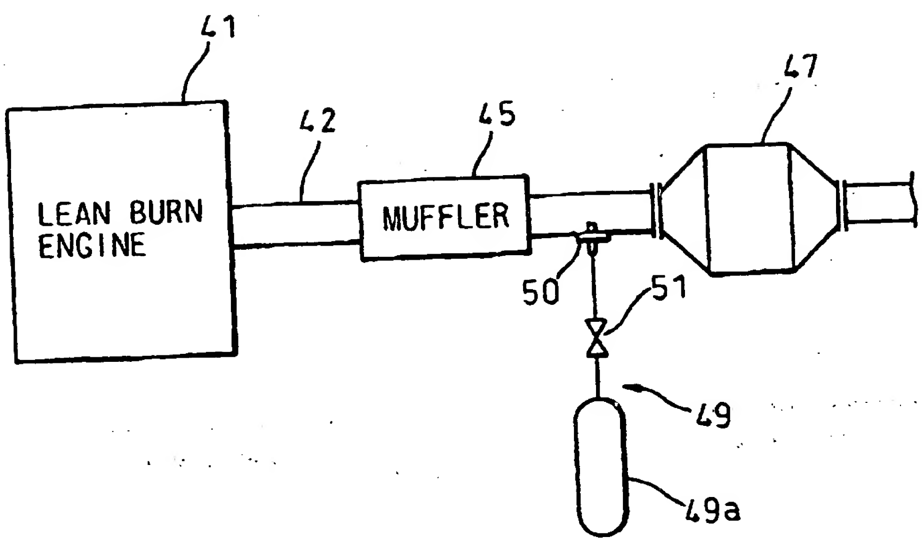
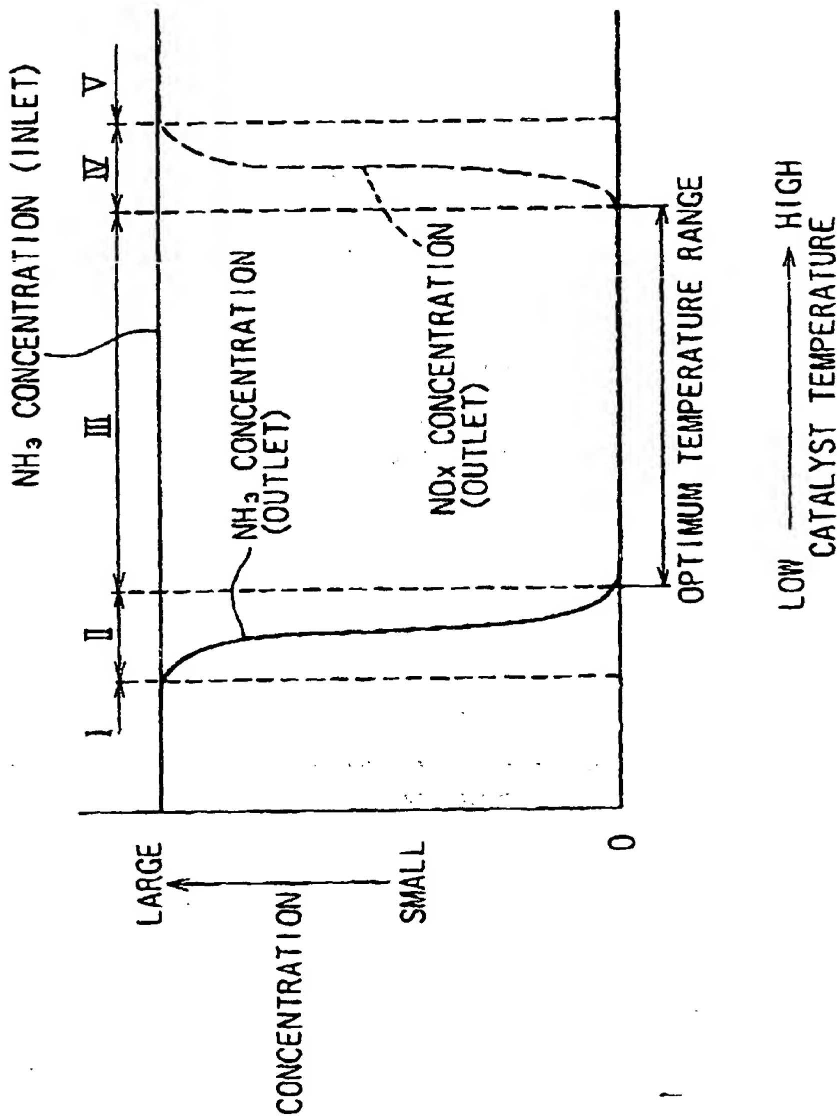


Fig.6


NH₃ CONCENTRATION (INLET)
I
II
III
IV
V
NH_3 CONCENTRATION (OUTLET)
NOx CONCENTRATION (OUTLET)
OPTIMUM TEMPERATURE RANGE
LOW → HIGH
CATALYST TEMPERATURE
CONCENTRATION
LARGE ← SMALL
0

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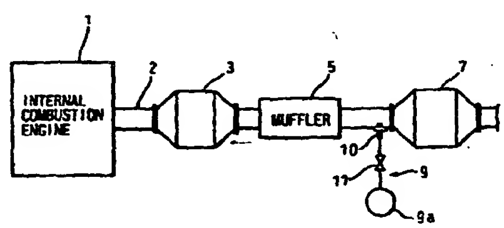
(54) A method for purifying combustion exhaust gas

(57) The method for purifying combustion exhaust gas according to the present invention utilizes a NH_3 decomposing catalyst. The NH_3 decomposing catalyst in the present invention is capable of converting substantially all of the NH_3 in the combustion exhaust gas to N_2 when the air-fuel ratio of the exhaust gas is lean and the temperature of the catalyst is within a predetermined optimum temperature range. Further, when the exhaust gas contains NO_x in addition to NH_3, the NH_3 decomposing catalyst is capable of reducing the NO_x in the optimum temperature range even though the air-fuel ratio of the exhaust gas is lean. In the present invention, the conditions of the exhaust gas containing NO_x are adjusted before it is fed to the NH_3 decomposing catalyst in such a manner that the temperature of the exhaust gas is within the optimum temperature range and the air-fuel ratio of the exhaust gas is lean. Further, NH_3 is added to the exhaust gas before it is fed to the NH_3 decomposing catalyst. Therefore, a lean air-fuel ratio exhaust gas, at a temperature within the optimum temperature range, which contains both the NO_x and NH_3 is fed to the NH_3 decomposing catalyst, and the NO_x, as well as the NH_3, in the exhaust gas is completely resolved by the NH_3 decomposing catalyst.

Fig.1



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